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# POTENTIAL IMPROVEMENTS IN MATERIALS ACCOUNTING FOR AN INTERNATIONALLY SAFEGUARDED FUELS REPROCESSING PLANT

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## Abstract

The effectiveness of improved materials accounting was evaluated using computer modeling, simulation, and analysis techniques for two model reprocessing plants. One plant, sized to 210 MTHM/yr, represents the small plants currently under international safeguards and the other, sized to 1500 MTHM/yr, represents the large plants expected in the future. The study indicates that conventional accounting may meet IAEA goal quantities and detection times for low-enriched uranium in these facilities. Dynamic materials accounting can meet the IAEA goal for detecting abrupt (1-3 wk) diversion of 8 kg of plutonium. Current materials accounting techniques probably cannot meet the protracted diversion goal of detecting 8 kg for plutonium in 1 yr. Facility design features that can improve the effectiveness of materials accounting in future plants are discussed.

## 1. Introduction

It is generally accepted that nuclear materials accounting procedures currently applied in operating nuclear reprocessing facilities will not meet more stringent international safeguards requirements for the high throughput facilities expected to be built during the last decade of this century. Improved materials accounting techniques were evaluated for (1) a present-day small (210 MTHM/yr) reprocessing plant and (2) a future large (1500 MTHM/yr) reprocessing plant. Considerations of process flows and plant designs has led to development of design criteria that can improve the effectiveness of materials accounting in future reprocessing plants.

The detailed terms and conditions for safeguarding specific facilities within nations signatory to the Treaty on the Non-Proliferation of Nuclear Weapons (NPT)[1] are negotiated with the International Atomic Energy Agency (IAEA) in accord with the general conditions of Article III of the NPT as set forth in the IAEA document INFCIRC/153.[2]

The objective of international safeguards, as declared by these documents, is the "...timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities... ." The emphasis is on "...the use of materials accountancy as a safeguards measure of fundamental importance, with containment and surveillance as important complementary measures... ."

INFCIRC/153, para. 31 also requires that the IAEA "shall make full use of the State's system of accounting for and control of all nuclear material subject to safeguards under the Agreement, and shall avoid unnecessary duplication of the State's accounting and control activities."

The application of international safeguards is negotiated between the IAEA (Agency) and the State (operator) on a case-by-case basis. "Goal quantities" for the detection of diversion have been proposed by the IAEA,[3] but are not presently a safeguards requirement. These "goals" are related to the quantities of nuclear materials required to produce an explosive device and the time necessary to convert these materials to that purpose. The goals include the detection of the diversion of:

- 75 kg of $^{235}U$ contained in low-enriched uranium over a period of one year.
- 8 kg of plutonium as nitrate or oxide in 1-3 weeks ("abrupt diversion").
- 8 kg of plutonium as nitrate or oxide over an entire year ("protracted diversion").

The Agency verification of the State's accounting system consists of three steps:

- Examination of the information provided in the Design Information Questionnaire and in subsequent routine and special accounting reports;
- Collection of independent information by the IAEA in inspections;
- Evaluation of the information provided by the State and collected in inspections for the purpose of determining the completeness, accuracy, and validity of the information provided by the State.

Inspection activity as defined in INFCIRC/153 permits approximately 3700 man hours (18 man years) and 1400 man hours (7 man years) of annual inspection, respectively, for plants having annual throughputs of 1500 and 210 MTHM. Thus, continuous inspection in such plants is foreseen.

## 2. The Reference Facilities

Using detailed process and measurement system modeling and simulation techniques,[4] various accounting strategies were evaluated for detecting diversion in a small (210 MTHM/yr) and a large (1500 MTHM/yr) reprocessing plant.[5,6] The PNC pilot facility at Tokai-mura, Japan, was studied as typical of plants currently under IAEA safeguards. The Allied-General Nuclear Services (AGNS) plant at Barnwell, South Carolina, was selected as representative of large facilities expected to be under IAEA safeguards in the 1990s. Both plants use conventional Purex technology to reprocess LWR reactor fuels having nominal plutonium concentrations of approximately 1%.

For this study, both reprocessing plants can be divided into four materials balance areas (MBAs).[7] The fuel receiving and storage area (MBA 1), the uranyl nitrate product storage area (MBA 3), and the plutonium nitrate product storage area (MBA 4) are shipper-receiver and item control areas. The chemical separations process area (MBA 2) is a process MBA in which periodic shutdown and cleanout physical inventories are augmented by near-real-time accounting techniques to improve the detection sensitivity and timeliness for short-term diversion. The accounting measurement strategies have been described previously.[5-9]

The chemical separations process area (MBA 2) can be treated as a single unit process accounting area (UPAA) for near-real-time materials balances, or it can be divided into multiple UPAAs, depending on facility design and process operating conditions.

Large Reprocessing Plant.

The MBA 2 for the AGNS facility was considered as a single UPAA (UPAA 1 2) and also was subdivided into two UPAAs: the codecontamination-partition process (UPAA 1) and the plutonium purification process (UPAA 2).

UPAA 1 2--Chemical Separations Process. The chemical separations process MBA is treated as a single UPAA (UPAA 1 2) by combining in-process inventory and flow measurements to form a dynamic materials balance approximately every 2 days. Under normal operating conditions, 2-1/2 accountability batches and 1 product batch are processed every day. Therefore, process logic suggests that a materials balance can be taken every two days to include an integral number of feed and product batches. Smaller batches, for example to high-level wastes, are included in the materials balances when the measurements become available.

Alternatively, a materials balance could be taken around UPAA 1 2 after each feed batch (approximately every 9.6 h) if on-line flow and concentration measurements are added to the plutonium product stream.

UPAA 1--Codecontamination-Partitioning Processes. A separate UPAA can be formed around the codecontamination-partitioning processes if flow-rate and concentration measurements are added to the intermediate plutonium product after partitioning and recycle streams. A dynamic materials balance can be taken about UPAA 1 for each feed accountability batch (every 9.6 h) by combining measurements of the concentration and volume of the feed batch, the concentrations and flow-rate in the intermediate, product, recycle, and waste streams, and the intervening in-process inventories in the process vessels.

UPAA 2--Plutonium Purification Process. Dynamic materials balances can be taken about the plutonium purification process if flow and concentration measurements are added to the aqueous and organic recycle streams. Materials balances can be taken using one of two product measurements: concentration and volume measurements in the plutonium product sample tank or integrated flow-rate and concentration measurements on the concentrator product stream. Contactor in-process inventory may be estimated using process operating data.[10]

Small Reprocessing Plant.

The MBA 2 for the facility was treated as a single UPAA (UPAA 1 2) but could be subdivided into three UPAAs: the codecontamination process (UPAA 1A), the partition process (UPAA 1B), and the plutonium purification process (UPAA 2). This further subdivision of the process is possible because buffer tanks installed between the codecontamination and partition processes permit additional transfer measurements between these processes. It is important to note that because of the low throughput of the Tokai plant, adequate sensitivity to short-term diversion is attained treating MBA 2 as a single UPAA; however, for larger facilities installed, suitably sized buffer tanks could improve the diversion detection sensitivity and localization.

## 3. Effectiveness of Materials Measurement Accounting

Table I lists Uranium-235 materials balance standard deviations in the process MBAs of the reference facilities. The materials balance standard deviations are based on a shutdown and cleanout physical inventory and were calculated using state-of-the-art measurement techniques. A range of values is given for two cases. In the first case, feed and product concentration measurements were recalibrated every 2 days. In the second case, there are no instrument recalibrations within the accounting period. These materials accounting sensitivities will be degraded if high-quality measurements cannot be obtained. Conversely, the sensitivities could approach the values of the first case if measurement errors can be controlled more effectively by identifying dominant error sources and establishing effective measurement control procedures. Note that the diversion detection sensitivity is ~3.3 times the materials balance standard deviation for a 95% detection probability and a false-alarm probability of 5%. From our analysis, we conclude that:

TABLE I

URANIUM-235 MATERIALS ACCOUNTING IN THE REFERENCE FACILITIES

| Accounting Period (months) | Materials Balance Standard Deviations (kg): Large Reference Facility Chemical Separations Area | Small Reference Facility Chemical Separations Area |
|---|---|---|
| 3 | 6.1-10.4 | 0.6-1.1 |
| 6 | 11.6-20.3 | 1.5-2.6 |
| 12 | 22.3-40.1 | 2.8-5.1 |

- For $^{235}U$ the proposed IAEA criteria for diversion sensitivity and timeliness probably are attainable by conventional materials accountability for the small reprocessing plant. For the large plant sensitivity criteria can only be met if rigorous materials measurement control programs are instituted.

Table II summarizes plutonium materials balance standard deviations for the process MBAs of the reference facilities. Additional accounting strategies are discussed in Sec. V of Ref. 6. Materials balance standard deviations for accounting periods ≤1 month are based on in-process inventory measurements while the process is operating. Materials balances standard deviations for accounting periods >1 month are based on a shutdown and cleanout physical inventory. In each case, a range of uncertainties is given for the entire process area. The cases considered range from best-case estimates of contactor in-process inventories with 2-day recalibrations of feed and product flow and concentration measurements to worst-case estimates of in-process inventories and no recalibrations within the accounting periods. Note that the diversion detection sensitivity is ~3.3 times the materials balance standard deviation for a 95% detection probability and an false-alarm probability of 5%.

In examining materials accounting sensitivities, we further conclude that for plutonium:

- In the <u>large</u> chemical separations process area, the proposed IAEA criteria for detecting <u>abrupt</u> diversion can probably be met if a rigorous measurement control program is undertaken.
- In the <u>large</u> chemical separations process area, the proposed IAEA criteria for detecting <u>protracted</u> diversion cannot be met; the goal quantity is only 0.05% of the annual plant throughput.
- In the <u>small</u> chemical separations process area, proposed IAEA criteria for <u>abrupt</u> diversion probably can be met.
- In the <u>small</u> chemical separations process area, the proposed IAEA criteria for <u>protracted</u> diversion may be achievable.

### 4. Process Design Considerations to Improve Materials Accounting

Features of process design and operation can strongly affect the application of materials accounting methods to high-throughput reprocessing facilities.[5,11,12] Traditionally, little consideration is given to safeguards effectiveness before the establishment of a plant and process design. Safeguards system designers are presented either with an existing facility or with a relatively complete and fixed plant design. With the increased recognition of the importance of safeguards, the situation may be

TABLE II

PLUTONIUM MATERIALS ACCOUNTING IN THE REFERENCE FACILITIES[a]

| | Large Reference Facility | | | | Small Reference Facility | |
|---|---|---|---|---|---|---|
| | Chemical Separations Area | | Conversion Area | | Chemical Separations Area | |
| Accounting Period | Materials Balance Frequency[b] | σ (kg Pu) | Materials Balance Frequency[c] | σ (kg Pu)[d] | Materials Balance Frequency[e] | σ (kg Pu) |
| 1 balance | 1/2 days | 2.1-2.4 | 1/2.88 h | 0.40 | 1/day | 0.25-0.38 |
| 1 day | --- | --- | 1/2.88 h | 0.43 | 1/day | 0.25-0.38 |
| 1 wk | 1/2 days | 2.5-2.8 | 1/2.88 h | 0.70-0.85 | 1/day | 0.30-0.43 |
| 2 wk | 1/2 days | 3.0-3.6 | 1/2.88 h | 1.1-1.4 | 1/day | 0.36-0.52 |
| 1 month | 1/2 days | 4.0-5.7 | 1/2.88 h | 2.0-2.4 | 1/day | 0.48-0.77 |
| 2 months | --- | --- | 1/2 months | 4.0-4.9 | --- | --- |
| 3 months | 1/3 months | 7.5-14.0 | 1/3 months | 5.9-7.2 | 1/3 months | 0.94-1.9 |
| 6 months | 1/6 months | 13.0-26.8 | --- | --- | 1/6 months | 1.7-3.7 |
| 1 yr | 1/yr | 23.8-52.7 | --- | -- | 1/yr | 3.2-7.3 |

[a]Materials balance standard deviations for accounting periods ≤1 month are based on in-process inventory measurements while the process is operating. Materials balance standard deviations for accounting periods >1 month are based on a shutdown and cleanout physical inventory. Ranges are given for the cases that are considered in Sec. V, Ref. 6. Unless otherwise noted, the accounting strategies shown here use chemical analysis techniques for the feed and product batches.

[b]Dynamic materials balances taken every 2 days include five input accountability batches and two product batches.

[c]Dynamic materials balances taken every 2.88 h include one input accountability batch and three product batches.

[d]An on-line measurement technique for the product batches is used in the accounting strategy shown here. This measurement is replaced by the result of chemical analysis of a sample ~8 h after the batch is produced.

[e]Dynamic materials balances taken every day (immediately after the product evaporator is drained) include two feed accountability batches and one product batch.

changing, and, in future plant designs, safeguards criteria should be regarded as equivalent in importance with health, safety, and economic considerations.

Guiding Principles.

Certain guiding principles govern effective materials control and accounting in any nuclear materials process. Each measurement is important for its impact on the loss-detection sensitivity. Thus, the necessity for each measurement point and the desired quality of each measurement should be judged by systematically analyzing the anticipated effects on materials-accounting sensitivity.

In high-throughput processes, the relative accuracy between feed and product measurements limits the long-term detection sensitivity,[13,14] and long-term relative biases between feed and product measurements should be controlled. Theoretically, the limiting factor is the uncertainty in the relative bias between the physical standards used for these measurements, which may be <0.1%. To approach this limit, sources of long-term measurement bias other than standards must be controlled by careful design of the sampling, measurement, and calibration hardware and procedures (Ref. 15, Part F; Refs. 16-19). Feed and product accountability vessels must be designed for accurate calibration and should be accessible for calibration checks and periodic recalibrations.

In dynamic materials accounting, the precision of the in-process inventory measurements and the variability of any unmeasured holdup are the limiting uncertainties in short-term detection.[5] The majority of the inventory should be in tanks and vessels that are instrumented for on-line measurements. Precisions of 1 to 5% generally are adequate. However, even with very precise measurements, large buffer-storage tanks may introduce large absolute errors that will seriously degrade the short-term detection sensitivity. On the other hand, relatively minor holdups and sidestreams will have little effect on detection sensitivity, and estimates based on historical data can be used until these components are measured, for example, during a physical inventory.

If all major in-process-inventory and process-stream components are measured, dynamic materials balances can be drawn around transfers between tanks and across vessels. Such balances may not require the precision and accuracy of conventional accountability, but they will be both sensitive and timely in absolute terms. Thus, process-design criteria for materials accounting should specify measurements for all the major inventory and flow components, and these criteria should be integrated early in the plant design.

Design Criteria for Future Reprocessing Facilities.

The materials accounting systems were studied for existing or designed facilities; therefore, safeguards criteria did not influence facility design. Several areas were identified where modifications in the plant or process design could improve the materials accounting system.

NDA on Spent Fuel. Nondestructive assay methods should be considered for verifying fuel burnup and/or fissile content of irradiated fuel in the fuel receiving and storage area.[20-22]

Input Accountability. The accountability tank and measurement procedures should be designed to provide the most accurate analyses and verification of nuclear material input to the accountability tank. If accountability tank heels and recycle material degrade the quality of isotope correlation analyses, provision should be made for obtaining samples directly from the dissolver. Recycle $HNO_3$ containing plutonium should not be used in the dissolver or accountability tank.

To minimize the effect of contained particulates, the centrifuge should be located ahead of the accountability tank.

Solvent Extraction Contactors. Uncertainties in contactor inventory have been identified as a limiting factor in short-term detection of diversion from the process area of reprocessing plants. Improved models of contactor operation would upgrade in-process inventory estimates.[23] Where practical, pulsed columns and mixer-settlers should be replaced with centrifugal contactors to minimize in-process inventory and to decrease the time required for obtaining a drain-down inventory.

Codecontamination Cycle. Codecontamination can be improved by increasing the number of stages in the HA contactor or by providing an additional decontamination cycle. The reduced radioactivity would facilitate plutonium concentration measurements at the input to the plutonium purification cycles.

Process Tanks. All process tanks containing significant quantities of nuclear material should be instrumented for liquid level, density, acid, and temperature measurements. These process data facilitate estimating the in-process inventory.

Process Buffer Tanks. Installing buffer tanks between major process areas decouples and thereby assists in defining UPAAs. Decoupling could allow process interruption in the plutonium purification area without disrupting head-end operations.

Product Concentration. Collocation of the reprocessing and conversion facilities eliminates the need for the plutonium product concentrator at the output of the reprocessing plant. The plutonium output concentration of the final plutonium purification cycle could be tailored to the requirements of the conversion process.

Process Stream Measurements. Flow meters capable of measuring flow rates to 1% or better in major process streams and 5-10% in waste streams should be installed. In-line or at-line detectors should be incorporated to measure plutonium concentrations to 1% or better in process streams and 5-20% in waste streams.

Instrument Accessibility. All instruments at key measurement points should be accessible for maintenance and recalibration. All sensors should be tamper resistant or tamper-indicating for use by international inspectors.

Redundant Instrumentation. Redundant flow, volume, and concentration instrumentation should be considered at KMPs.

Computer Data Handling. All data acquisition and analysis should be performed by computer, and all in-line and laboratory instruments should be linked to the data-base computer.

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